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Advanced Technology for Radium Removal from Drinking Water
The Flatonia Water Treatment Project

Texas State Dept. of Health, Austin

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16. ABSTRACT

A study was conducted to determine the applicability of using manganese-coated acrylic fibers to remove Radium from drinking water.
The technique of using Manganese-coated fibers to remove radium from drinking water is a simple, effective and economical procedure which is compatible with the operations of a municipal utility crew.

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Division of Occupational Health
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Advanced Technology for Radium Removal from Drinking Water: The Flatonia Water Treatment Project

Lewis M. Cook

Texas Department of Health

I. Introduction

In late 1977, the Texas Department of Health received a grant from the U.S. Environmental Protection Agency (EPA) to conduct a study to determine the applicability of using manganese-coated acrylic fibers (Mn-fibers) to remove radium from drinking water. The project also had as its goals the reduction of the levels of radium-226 in the municipal water system of Flatonia, Texas. Flatonia (Figure 1), a small (population 1,108) town on Interstate 10 approximately mid-way between Houston and San Antonio in southeast Texas, depends entirely on ground water (well water) for the municipal supply. Three of the four wells in the water system exceeded EPA's limit of 5 picoCuries total radium per liter (pCi/l) for drinking water and two of those exceeded it substantially. About the time the project was begun, the city drilled another well to augment the system because the well having the lowest level of radium had failed.

During the course of the project, some of the objectives had to be changed, due to changes in water useage, philosophy, and waste-generating and disposal requirements.

One of the goals was to remove the radium from the filters so that the filters could be re-used. This was not attempted for several reasons. The removal of radium requires that the filters be immersed in Nitric acid, and the acid, containing fairly high dissolved and suspended radium concentrations, would require disposal in liquid form. Since waste disposal facilities no longer accept liquids for disposal, and

Figure 1. Business district, North Main Street, (U.S. 90), Flatonia, located in Fayette County, Texas.

the fact that it was thought that the handling of large volumes of liquid radioactive waste was not a task compatible with the operations of a municipal utility crew, this part of the proposed project was not attempted.

Despite the many problems encountered, problems which ranged from tangled red tape and logistics to bad luck that could only be termed bizarre, and despite the numerous delays and modified objectives, a great deal of information about the process was gained and the project was technologically successful. The water supply in Flatonia met the drinking water standards for radium for almost all of 1979 for the first time in several years.

From the knowledge gained performing this work, we have concluded the technique of using Mn-fibers to remove radium from drinking water appears to be a relatively simple, effective procedure which can be compatible with the operations of a municipal utility crew and probably will operate at an economic advantage over other methods.

II. History

The Texas Department of Health began routine sampling of water near uranium deposits in 1970. Private wells near these deposits have been found to deliver water containing over 100 pCi/l of radium-226 to the families using this water. The work was first reported at the Natural Radiation Environment II Symposium (Wukasch and Cook, 1972), and was further discussed at an International Radiation Protection Association Symposium in Washington, D.C. (Wukasch and Cook, 1973).

A Washington Post story summarizing the paper came to the attention of W.S. Moore, Ph.D., who contacted us.

Moore, at the time, was working with the U.S. Navy Oceanographic Office, and was examining the ratios of radium-226 to radium-228 in the oceans to determine oceanic vertical mixing rates. Because the concentration of radium in ocean waters is quite low (on the order of 0.05 pCi/l) it was necessary to concentrate the radium from large volumes of sea water in order to have sufficient quantities of radium to determine this ratio. The removal was done with acrylic fiber filters which had been treated with manganese solutions. Radium removal efficiencies were on the order of 90% (Moore and Reid, 1973). Within a month, Moore had brought an experimental apparatus to Texas at our request, to determine if the process would reduce the high levels of radium being found in some well water. It did (Moore and Cook, 1975).

About the same time, we were notified that gamma ray well logs of wells in

Flatonia showed extremely high levels of gamma ray intensity in the (Miocene or Oligocene Series) Catahoula tuff (Hill, 1973).

The Catahoula tuff is a host rock for uranium deposits in other parts of South Texas (Cook, in press), and uranium exploration in the near vicinity of the town had been conducted. No plans for commercial operation had been announced, however.

Water sampling showed that all of the wells in the Flatonia municipal water supply contained detectable levels of radium and radon (Table I). Well locations are shown on the map of the town in Figure 2.

In August of 1974, Moore and the writer performed tests using the process on water from well #6 in Flatonia, similar to those done in South Texas in 1973 (Figure 3). In February 1975, other variations of the process were also tested with encouraging results.

In order for the city water supply to meet the drinking water standard, it would be necessary for the city to either abandon the present wells and drill new wells, gambling that those wells would have acceptable radium levels, or the city could treat the water from the wells to reduce the radium content.

When the costs of drilling new wells were compared to the projected costs of treating the water by passing it through Mn-fibers, it was decided that a potential for substantial savings lay in using the fibers.

The process was an experimental one, and if it could successfully be scaled up to water-treatment plant size, then other communities which have problems

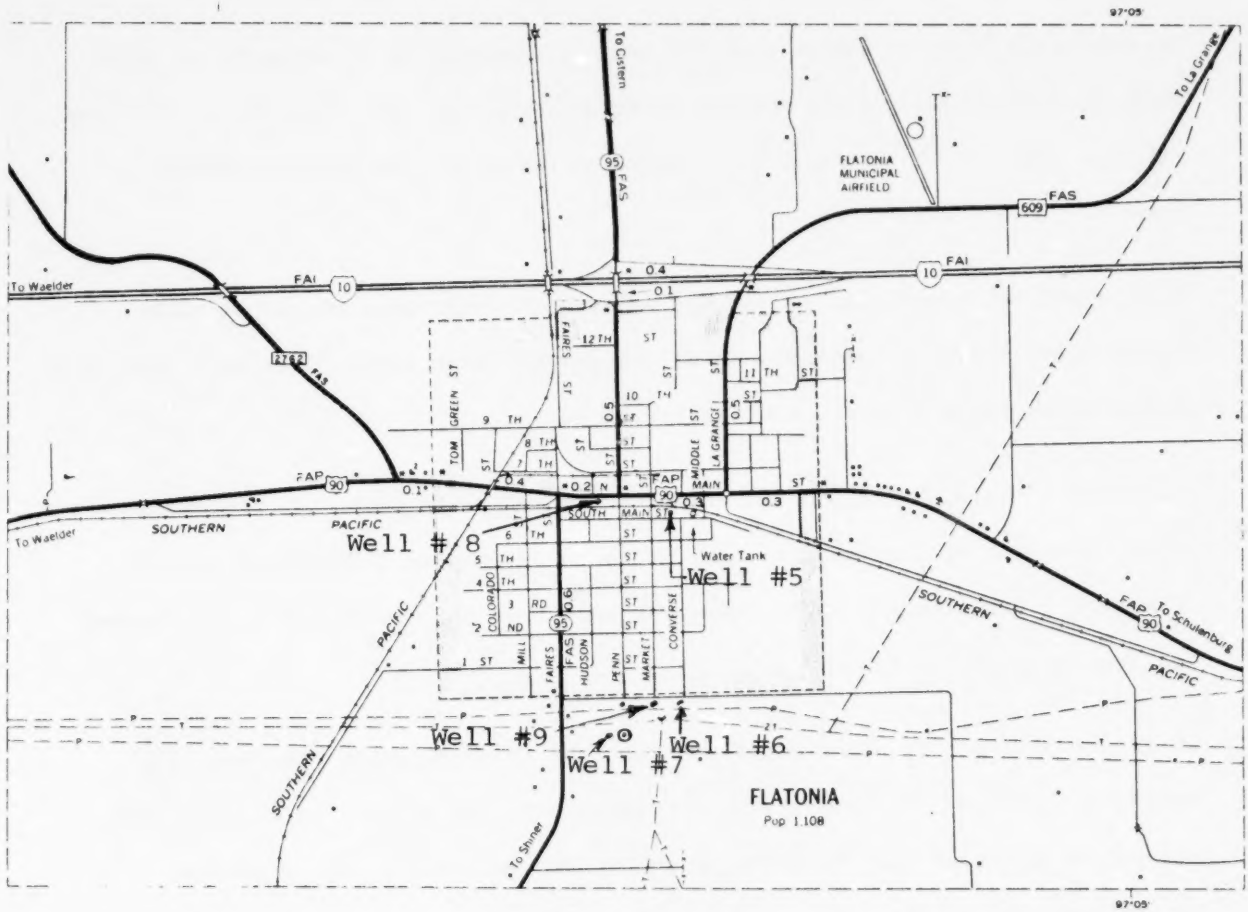

Figure 2. Well locations in Flatonia.

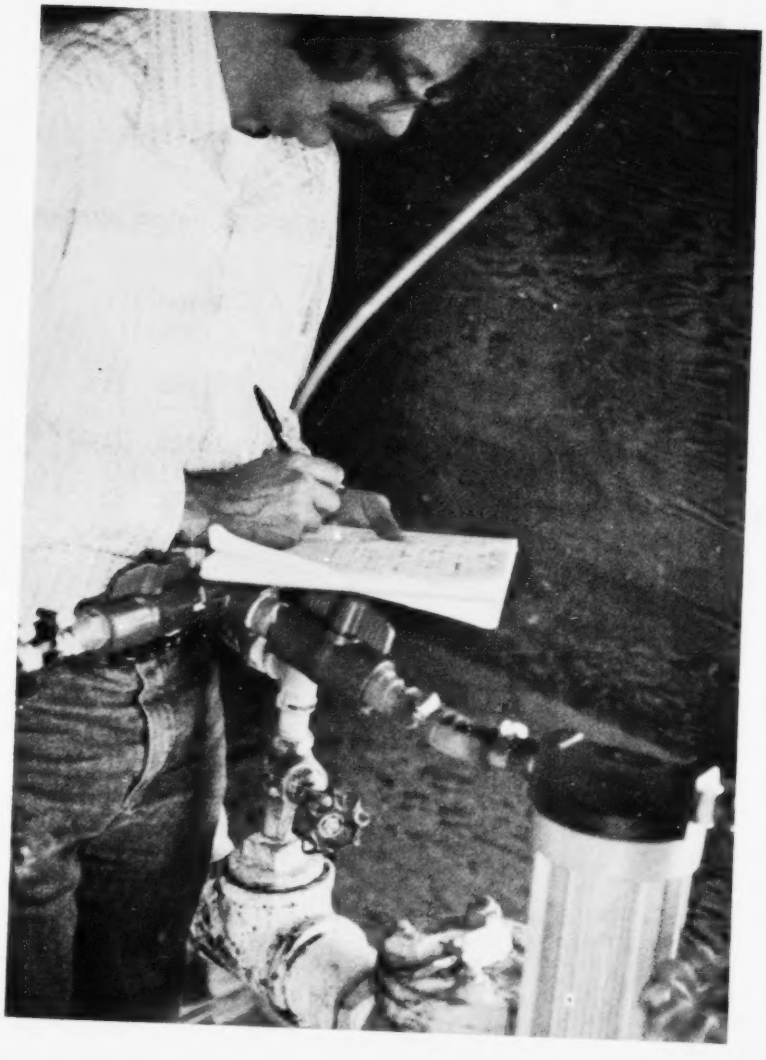

Figure 3. Bench scale tests of the process were conducted in Flatonia using 10 inch length filter cartridges. 30 inch length filter cartridges are used in the housing in Flatonia.

Table 1

Radium and Radon in the Flatonia Municipal Water System

	Radium-226 (pCi/l)	Radon-222 (pCi/l)
Well 5	3.4 (3)	1300 (1)
Well 6	16 (13)	10700 (1)
Well 7	14 (4)	7400 (1)
Well 8	6.4 (3)	1400 (1)

Numbers in parentheses are number of samples.

with high radium levels in their drinking water would berefit from this new technology.

It was thought that development of such a process would logically be of interest to the EPA, so contacts with EPA were made and in September of 1977 a proposal for the Flatonia Water Treatment Project was submitted. A cost-reimbursement contract was subsequently issued.

The proposal entailed participation of three governmental entities: EPA, the Texas Department of Health, and the City of Flatonia. The EPA provided money for the purchase of equipment and supplies, consultation fees and interstate travel. The Health Department paid the salaries of the principle investigator, his assistant, and paid for laboratory services and intra-state travel. The city of Flatonia furnished the work space, people and equipment for installation of process equipment and sample collection as well as work space in Flatonia. W.S. Moore, who holds U.S. patent numbers 3,965,283 and 4,087,583 on the fibers and their preparation, allowed this technology to be utilized on a royalty-free basis.

III. Operations Conducted

The Mn-filter preparation area was constructed in the fall of 1977 in anticipation of using the area as a filter regeneration facility. Accordingly, an emergency shower was also installed, as was an extra large capacity (about 2,000 liters) spill absorption and containment "tray" in the bottom of the work area. Vermiculite was used as the absorber. (Figure 4)

The filter housing foundation for the south plant area was placed near the buried water lines leading to the elevated ground storage tank on the south edge of town. (Figure 5). The lines from well 6 and well 7 connect just to the west of this area.

Well 9 was drilled and developed and was connected into the vertical riser pipe at the tank. This routing was chosen so that the water from wells 6 and 7 could be routed through the filter and the water from well 9 could be put into the system without being sent through the filter. The pipe from well 9 to the tank crosses the feed pipe from well 6 near well 9, and had the radium removal equipment not been in the system, there would have been a connection at the point where the well 9 output pipe crosses the well 6 pipe. The length of pipe necessary to extend the well 9 connection to the tank was supplied to the city under this project.

The filter housing was installed in the Flatonia system about one year after the foundation was prepared, due to difficulties discussed elsewhere in this report.

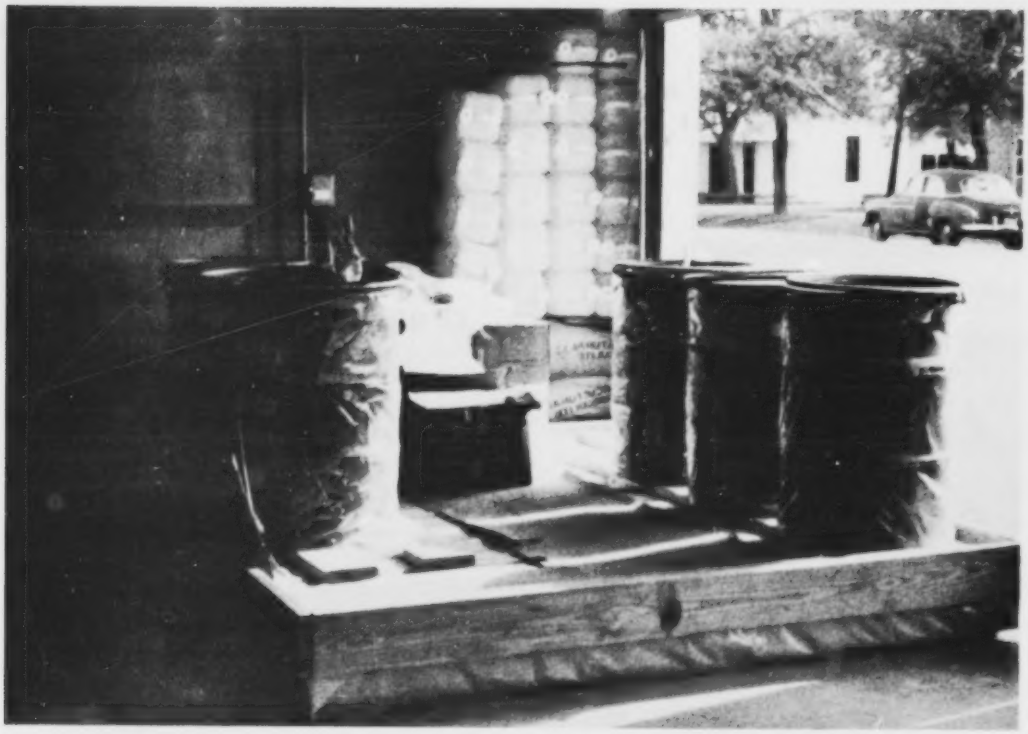

Figure 4. The filter preparation area was to be used for filter preparation and regeneration. The use of 2 x 8 lumber provided a sturdy platform for the drums, which weigh about 500 pounds each when full.

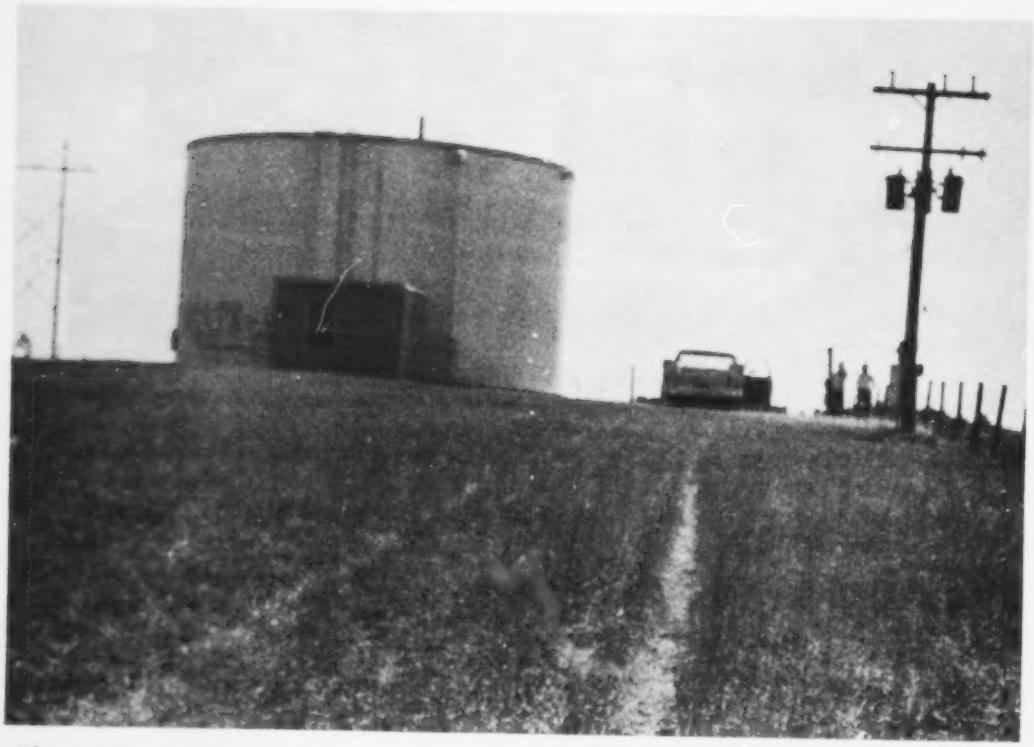

Figure 5. Location for the South Water Treatment Plant.

The foundation (6' x 6' x 9") was constructed in December 1977 of reinforced concrete (Figures 6 and 7). This proved to be larger than strictly necessary but provided a comfortable work area.

After the filter housing arrived, the plumbing was connected. Difficulty in cutting cast iron pipe was encountered. Since the mechanical joint pipe fittings required nipples between adjacent fittings, small pieces of the pipe were needed. A hydraulic pipe cutter (Figure 8) was used, but failed to cut the pipe. The pipe, instead of fracturing like case iron does, merely deformed under the pressure. The pipe was supposed to have been cast iron but a well-intentioned contractor had substituted a stronger, less brittle pipe.

The city crew supplied a small piece of cast iron pipe for some of the needed fittings and the mild steel pipe was used for the rest. The city crew took the pipe to Prototype Machine Shop, where it was cut into lengths using a band saw (several bands were used in the attempt). No charge was levied for this service.

The city crew also dug up the buried water line and made the hook-up.

In the meantime, filters had been treated. Figure 9 shows the beginning steps and Figure 10 shows 60 treated filters. The surgical gloves used to protect hands proved to be not sufficiently durable. Stains from the permanganate last a few days to a week before they wear off. The solutions, when spilled on the skin, do not cause pain (Figure 11).

The filter housing hardware was installed (Figure 12), the filters inserted, and the spring and cap assemblies were joined and pushed onto the filter tops (Figure 13). Next the hold-down plate was installed, the head bolted down,

Figure 6. Constructing the form for the foundation of the South Water Treatment Plant.

Figure 7. The finished form.

Figure 8. Problems were encountered in cutting pipe supplied under contract. The supplied pipe was not cast iron but a more durable and stronger, mild steel. The hydraulic pipe cutter operates only on brittle pipe.

Figure 9. Filters at initial chemical addition step.

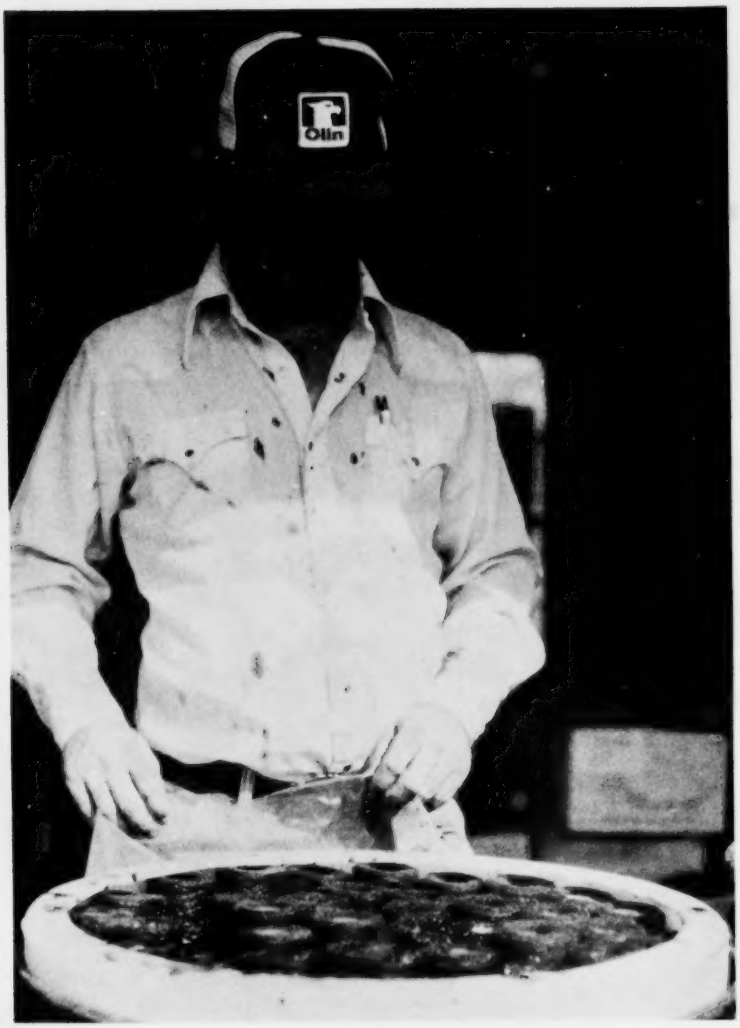

Figure 10. Filters at end of treatment.

Figure 11. Manganese stains result when holes develop in gloves. The stains cause a short-term discoloration, are painless and should remove radium from water.

Figure 12. Installation of "V" shaped filter guide bars. Note circular hold-down plate which is used to compress filter springs.

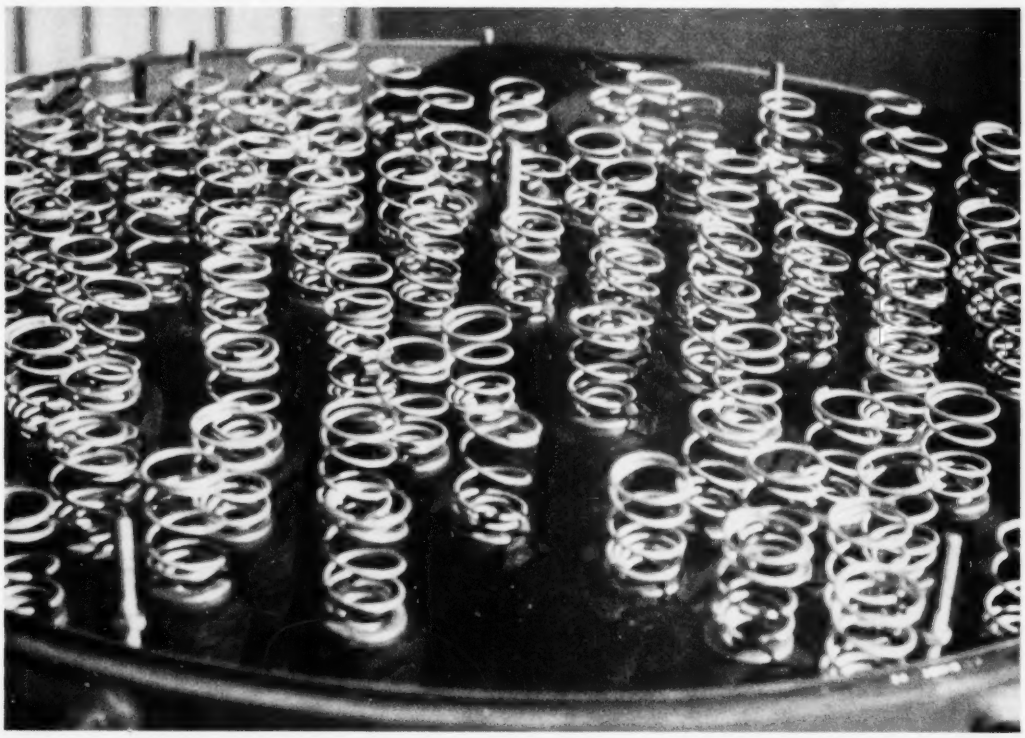

Figure 13. Spring and cap assemblies are inserted into the top of the filters. Care must be exercised to avoid knocking these assemblies over when the hold-down plate is employed.

and testing was begun (Figure 14).

Tests for chemical changes in the water before and after it passed through the filters included radium removal, presence of acrylonitrile and/or other organic chemicals as well as routine chemical analyses and taste and odor (Table II & Figure 15). The treated water was dumped on the ground during this early data gathering period between the initial installation and receipt of laboratory results indicating acceptable quality water was delivered.

Prior to the beginning of the tests, cards were mailed to the customers of the utility, briefly describing the project and requesting notification of any complaints. A few complaints were received concerning aesthetic quality of the water prior to the time that any water was treated by the filtering system. After the system was connected, no complaints related to the operation were received. The card is shown in Figure 16.

The radium removal efficiency was substantial, but less than expected. As a result, the treated water was added to the system only when needed after the initial trial run.

In June 1979, the filters were exchanged with a new batch to test several theories about the process, which will be discussed in detail later in this report. At that time, most of the interior of the filter housing were painted with epoxy paint.

The second filter housing is to be installed at the downtown water plant (Figure 17). The city warehouse just to the west of this plant is shown in Figure 18. It is in this building that our filter preparation area is housed.

Table II

Chemical Tests - well 6 water

Test	Before Filter	After 1000 gal.	After 4000 gal.	After 8000 gal.
Calcium	108	88	92	101
Magnesium	4	4	3	4
Sodium	67	61	60	59
Carbonate	0	0	0	0
Bicarbonate	346	336	337	341
Sulphate	24	23	23	24
Chloride	81	79	80	73
Fluoride	0.4	0.4	0.4	0.4
Nitrate	< 0.02	< 0.02	< 0.02	< 0.02
Potassium	NA	29	24	NA
Dissolved Solids	454	449	448	429
Phenolphthalein Alkalinity as $CaCO_3$	0	0	0	0
Total Alkalinity as $CaCO_3$	284	275	276	280
Total Hardness as $CaCO_3$	285	233	243	271
Diluted Conductance (µmhos/cm)	870	864	864	852
pH	7.6	7.5	7.6	7.7

Gas Chromatography + Mass spectroscopy test for acrylonitrile and related chemicals negative.

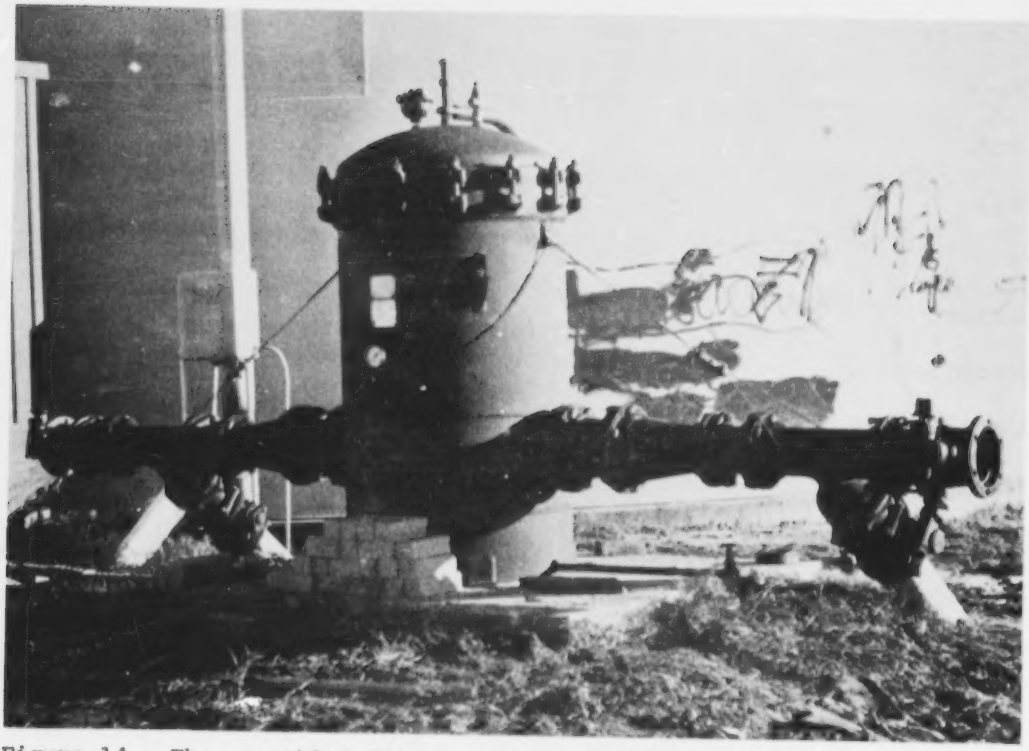

Figure 14. The assembled filter mechanism at the early testing stage.

Figure 15. Tests for chemical, radiological, and aesthetic quality were conducted prior to adding water to the municipal water system.

NOTICE

The Texas Department of Health, working in cooperation with the City of Flatonia with financial assistance from the U. S. Environmental Protection Agency, will begin a new water treatment process in a few weeks. After preliminary testing, the water from some of the City's wells will be passed through specially treated filters before being added to the distribution system. The filters are spun from acrylic yarn and treated with oxides of manganese. They will remove radium, naturally present in some of the well waters, without changing the appearance or taste of the water.

No problems with the delivered water are expected, however, if you should have a complaint, please save a sample of the water and call the City Office at 865-3337.

Figure 16.

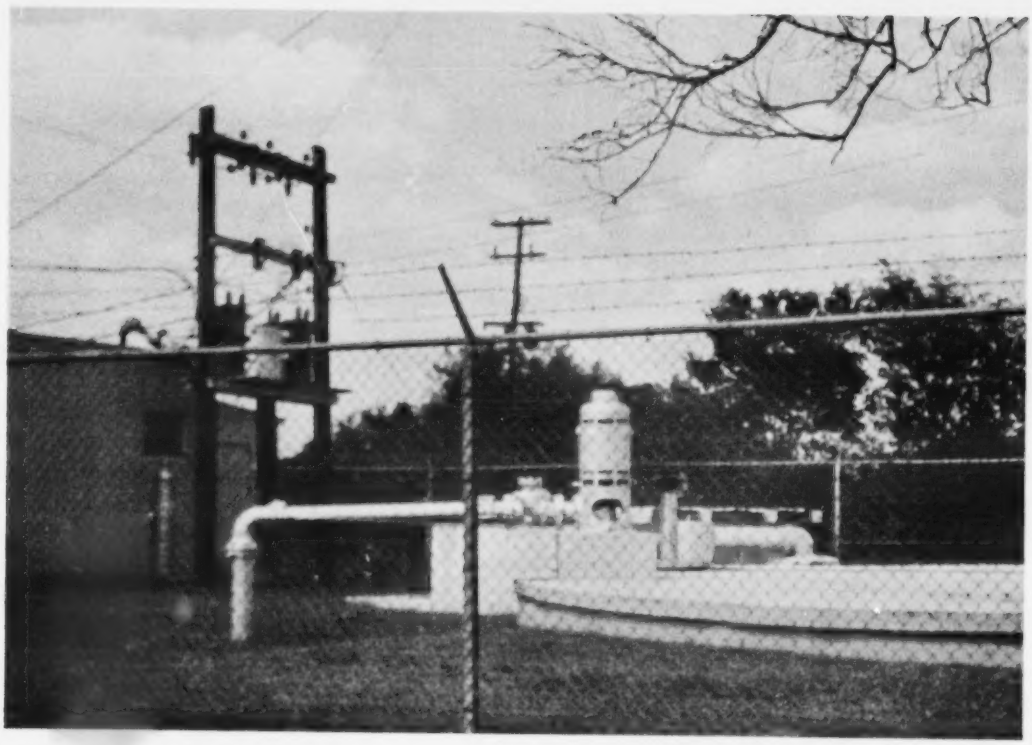

Figure 17. Downtown water treatment area where second filter unit is to be installed.

Figure 18. The City Warehouse is shown to the left of the base of the elevated water storage tank. It is the location of the filter preparation area.

Forms for this second unit foundation have been made, and this will be smaller than the foundation for the first unit. The city will conduct the operation of this second installation and filter operation under guidance and consultation with the Health Department.

IV. Logistic Problems Encountered

A. Delays

The project was designed to last one year, based upon expected paperwork delays, manufacturing schedules, available travel money, and manpower.

Delays far exceeding reasonable expectations were encountered. Reviews of purchase orders for the filter housings by the Fiscal Division of the Health Department, followed by a review and bid requests issued by the Texas Board of Control, bid opening , and the award of contracts required from 8 - 11 weeks.

Construction of the first filter housing took 22 weeks, although 6 - 8 weeks had been the estimate given by the supplier.

That first filter housing actually never made it to Flatonia. It was destroyed in shipment when vandals burned the shipping warehouse in Austin on July 27, 1978, as the housing was awaiting forwarding to Flatonia. In the meantime, we had ordered the second housing. We planned to substitute it for the first housing. This replacement for the first filter housing didn't arrive in Flatonia until December 1978. In early February 1979, the paperwork for the purchase of the second filter housing was begun. The delivery of this second housing occurred on November 16, 1979. Installation of this second unit is planned by the Flatonia crew in the spring of 1980.

B. Materials

Several problems related to materials were encountered during the project. The treatment of the first set of filters proceeded slowly, much more slowly than expected. Two 55-gallon drums were filled with 60 filters for the treatment procedure. The drums were lined with polyethylene bags of 6 mil thickness, which proved to be insufficiently strong to prevent leaks and subsequent corrosive attack of the steel drum. Next, for protection of the drums they were coated with an epoxy paint used for water tanks. The paint, after drying more than the recommended curing time, was also attacked by the potassium permanganate. Finally, drum liners made of 1/8 inch thick polyethylene were lent to us by W.S. Moore, Ph.D., the consultant, in order that they could be obtained without significant delay.

The filter treatment consists of soaking the filters in a potassium permanganate solution for lengths of time which depend upon the strength of the solution and temperature (U.S. Patents 3,965,283 and 4,087,583). This requires about 7 - 10 minutes at 175°F and a concentration at 60 grams of potassium permanganate per liter of water. At 86°F, the processing requires 3 to 4 days, at the same solution strength.

The fibers, in becoming coated with oxides of manganese, turn from pure white through tan to a chocolate color (at about 4% manganese by weight) and finally to jet black (at around 10% manganese by weight). The desired degree of treatment is about 7 - 8% manganese by weight, resulting in a very dark drown fiber.

On or about March 7, 1978, before the filters were delivered, a phone conversation was held with the contractor, who was informed of the need for acrylic

fiber filters, and that any other substance would not work. The contractor stated that he had acrylic fiber filters and another type, a polyurethane, in stock, that the polyurethane was slightly more expensive and it would be supplied at the same price, if desired. This suggestion was rejected, again with the admonition given that only acrylic fibers were acceptable.

On March 29, 1978, 42 boxes of 12 filters each were delivered to Flatonia. The filters were inspected and appeared acceptable, being the correct length and diameter, being made of white fibers spun onto 1" plastic centers.

Immediately, 60 filters were placed in a drum containing a solution of potassium permanganate (40 g/l).

On March 31, a check on progress of the filter treatment was conducted. The temperature of the unheated drum was 72°F and the degree of manganese loading on the fibers was estimated to be less than 1%. On April 5, a drum heater was connected to a second drum filled with water and another 60 filters to determine heater settings.

On April 6, heaters were connected to both drums and chemicals were mixed for treatment of all the 120 filters, enough for one filling of the filter housing plus five for testing.

On April 7, the manganese level of the filter was checked. Again less than 1% manganese by weight was deposited on the filter. Because of possible interference caused by the epoxy coating on the drums by the chemicals, processing was suspended by draining the chemical solution and leaving the filters to stand in water.

Upon receipt of loaned polyethylene drum liners, processing was resumed on May 2, 1978.

Then, 60 of the filters were placed in a drum with about 20 grams of $KMnO_4$ per liter, heated to 83°F. On May 4, the temperature was increased to 93°F by Flatonia City workers. On May 5 the fibers had 1 - 2% manganese by weight, estimating from the color. The solution strength was increased to 40 grams per liter and the temperature increased to 98°F to speed the seemingly slow uptake of manganese.

By May 8, the fibers still had not absorbed more than 2% manganese by weight, so the solution strength was increased to 60 grams per liter, the "reference" strength, and the temperature was kept at 98°F.

On May 9, the city recorded the temperature as being 103°F, and it was left at that warm temperature until May 12, by which time it was anticipated they would have been treated to 7 - 10% manganese by weight.

The fibers actually contained less than 3% manganese. The drum heater was disconnected. A sample of 100% orlon acrylic (white) yarn was obtained to compare it with those being used in Flatonia. Part of one of the unused Flatonia filters was unwound and 3.8 gram bundled sample of each type of yarn was taken. They were dropped simultaneously in a heated solution of potassium permanganate solution at approximately 50 grams per liter at 180°F. They were stirred intermittently and removed together after 7 minutes in the hot potassium permanganate solution. They were then placed in a beaker and washed until no trace of the purple solution remained.

Because of the differing winding of the yarns, there was no possibility of confusing the two samples.

The orlon acrylic yarn reacted exactly as expected, turning jet black. The sample from the filter delivered to Flatonia reacted differently.

The degree of darkening on it was very much less than the orlon acrylic. The final color on this sample was beige, representing much less than 1% manganese by weight.

The consultant for the project, Willard S. Moore, Ph.D., the developer of the treatment process, was called and he reported having tested numerous acrylic fibers manufactured under differing processes and had never found an acrylic fiber which failed to take up manganese oxides.

As a final test, the vendor's filter material was heated to between 170°F and 190° F in the same solution used for the simultaneous test for over one hour and did not darken significantly more.

It was therefore concluded that the filters delivered to Flatonia were something other than the spun acrylic fiber filters specified. Laboratory tests for infrared reflectance spectrometry confirmed the discrepancy.

C. State Budgetary Process

It should be briefly mentioned that these were significant delays due to the somewhat complex purchasing system the State of Texas uses. No "petty cash" system existed, so for relatively small items which were needed quickly (in a few minutes rather than several days or weeks) the investigators purchased these with personal funds. Examples include extension cords, small springs, and bleach (for disinfecting).

V. Water Treatment

A. Radium Concentration Variations

Earlier testing of the activity of well 6 gave consistent results of around 14 pCi/l. During this earlier period, well 6 provided most of the water used by the town. At that time the town used wells, 6, 8, 5, and 7 for their supply, approximately in that order when volume is considered. Well 5 failed in 1977 and another well was drilled, well 9A. This new well did not produce the quantity that the driller had guaranteed, and another well, numbered 9B, was drilled by off-setting well 9A. This offset produced over 250 gallons per minute, a greater production rate than any other well in the system. It plus well 8 is sufficient to provide most of the water needed by Flatonia all during the fall, winter, and early spring, but demand during the summer requires all of the wells in the system.

When well 6 was turned on at the beginning of the project in December of 1978, the concentration of radium in the water was 38 pCi/l. Heavy pumping caused the concentration to drop to 20 pCi the next day. A similar pattern of activity was seen in June of 1979 after the second set of filters was installed and was used for treating the town's water in the summer.

Apparently, the most highly contaminated water is drawn from near the well, and water more distant from the well has less activity. After the well is shut down, the radium levels climb once again to around 35 -

40 pCi/l. After continued heavy use, the radium levels decreased to around the levels originally measured, around 14 pCi/l.

Well 7 showed a little variation also. The levels of radium in well 7 seemed to increase slightly with pumping.

B. Description of Filter Housing

The filter housing used in this project is a Facet Model 115R3, a cylindrical steel pressure vessel about 36 inches in diameter and standing about seven feet tall. The filters are 30 inches tall, and are arranged in a concentric circular pattern, with a "wedge" of filters missing on the outer rows to accommodate the inlet. The filters fit over "cups" with steel "V" channel guide bars inserted to hold the filters upright. The guide bars are removable and fit very loosely into the cups. The filters are held upright and vertical by a spring and cap assembly fitted into the top of the filters. A ½ inch thick circular steel plate, about 2 inches in diameter less than the inside diameter of the vessel, is bolted down over the springs, the springs being compressed, thereby holding the filters in place. This hold-down plate has a circular hole which is aligned above the intake. The doomed top of the housing is held on with swing bolts and sealed by a rubber "O" ring. The vessel is rated for 150 psig, with a maximum differential of 75 psig between the inlet and output.

The water flows into the housing at a level below the bottom of the filters, and rises in the empty "V" space above the inlet. From this point, it can pass among the filters and be filtered, or can rise and flow through the springs and down between the filters. Alternatively, the incoming water could flow up through the hole in the hold-down plate, then around the edge of the hold-down plate to the area between the tops of the filters and the plate, in the space where

the compressed springs stand.

The water passes through the filter, from the outside into the center hole of the filter and then down through the tube and through the 115 holes in the bottom of the filter chamber. The outlet for the housing is opposite the intake. The housing has a drain pipe on the bottom, which can be used for sampling, as a "bleed" or as an alternate output at relatively low flow rates (less than 150 gallons per minute), permitting dumping of the filtered water.

C. Results and Problems

The initial filter loading and tests in early December 1978, achieved a large reduction in the levels of radium in the water passed through the filter. Treated water was dumped on the ground until analysis results were received from the Health Department laboratory which showed an absence of any organic chemicals from the filtered water which could have come either from the manufacturer of the acrylic fiber of the chemical treatment in Flatonia. Even after heavily concentrating the sample, no traces of any chemicals attributable to the filter could be detected using gas chromatography and mass spectroscopy.

After the negative report on the presence of organics was received, and analyses for radium showed that radium levels were below 5 pCi/l for the commingled water, the filter output was put into the system.

Apart from this early work to prove that the process worked, the filtered water was added to the system only when it was needed to keep up with demand.

Well 9 was out of service for a few weeks in January 1979 and wells 6 and 7 were needed. During this period the 250,000 gallon elevated ground storage

tank needed cleaning of the sand and iron sulfide precipitate in it and well 6 was used to flush and fill this tank.

The reason for using well 6 as little as feasible was the fact that the efficiency of the filter system was not as great as had been expected, and adding the water that passed through the filter system to the municipal water supply when it wasn't strictly necessary would be contrary to the ALARA* principle.

Simply dumping the filtered water on the ground was also viewed as wasteful and was not done except for flushing loose manganese oxides from the filter system and for the initial testing phases.

The results of the water treatment project with respect to the removal of radium from the drinking water in Flatonia are shown in Table III.

The initial removal efficiency with the first set of filters was less than expected. Earlier work with smaller filters (although with larger flow per filter) had led to the expectation that initial removal efficiencies approaching 99% could be expected. The initial radium removal efficiencies were actually around 80%. The cause of this low efficiency was speculated to be due to one of several factors:

1. Possibly uneven manganese uptake by fibers in the middle of the winding of the cartridge's core.
2. "Channeling" through the filter housing.
3. Chemical Interference caused by Iron oxides (rust) in the filter housing.
4. An empty filter space in the housing.
5. Insufficient amount of manganese treatment of the fiber.

*As low as reasonably achievable.

Table III

Water Processing Results

Date	Time	Volume (Gal. x 1000)	Well	Radium Concentration Before Filters pCi/l	Radium Concentration After Filters pCi/l	Radium Removal %
12/05/78	1500	1	6	38	6.7	83
		4	6		6.6	
		8	6		3.6	
		16	6	28	4.0	86
12/06/78	1000	114	6	24	4.8	80
	1230	123	6	19	7.0	63
	1330	129	6	20	5.9	70
	1530	141	6	20	5.9	70
12/11/78	1205	255	6	26	7.1	73
	1305	261	6	22	6.3	71
	1405	267	6	22	7.1	67
	1505	273	6	23	7.2	68
12/13/78	1406	333	6	27	8.7	68
	1416	334	6	33	7.6	77
	1446	337	6	25	6.8	73
12/14/78	0880	409	6	(27)	8.2	70
	0950	535	6	(27)	4.6	83
	0950	535	6	(27)	4.6	83
12/18/78	unk	(877)	6	(27)	4.4	84
12/19/78	0745	991	6	(27)	5.8	78
12/20/78	0845	1123	6	(27)	9.4	65
12/21/78	0935	1249	6	29	7.0	80
1/18/79	1300	1252	6	32	10.9	66
	1340	1256	6	40	10.8	72
1/19/79	1050	1278	6+7	17	7.5	56
	1230	1290	6+7	19	6.1	68
	1330	1304	6+7	20	5.7	72
2/06/79	1320	1962	6	37	8.1	78
	1335	1964	6+7	32	6.9	79
	1350	1965	7	17	0.2	98
2/13/79		1966	7	9.5	2.0	79
6/22/79	1055	3	6+7	27	7.9	71
	1120	6	6	27	4.2	84
	1140	8	7	10	2.1	79
	1300	17	6+7	22	3.2	85

Date	Time	Volume (Gal. x 1000)	Well	Radium Concentration Before Filters pCi/l	Radium Concentration After Filters pCi/l	Radium Removal %
7/11/79	1300		6	19	5.1	73
	1305		7	18	7.0	61
	1310		6+7	25	1.1	95
8/09/79	1255	2468	6	16	6.5	59
	1320	2470	6	13	8.3	36

-1-

To test the first possibility, one of the unused filter cartridges was sacrificed to examine the interior windings of the fiber. The center tube, a one-inch diameter perforated tube, allows the solutions access to the interior windings, while access to the outside windings is obtained from being in contact with the solution in the drum, but the chemical solution can reach the windings in the middle of the filter only by diffusion from the center tube and from the outside.

There was no apparent difference in the manganese content of the fibers in the middle of the cartridge compared to the inside windings or the outside windings.

-2-

Based upon data gathered from radiation surveys, channeling was suspected. A method to test this supposition was devised. Channeling of the water through the filter housing, causing some filters to filter more water than others, would mean that the water would move more quickly through some of the filters. This, in turn, would mean that the water would spend less time in the filter medium and a smaller percentage of radium would be removed.

The filter housing is designed so that it is likely that some of the filters would have less flow through them than others. The incoming water is routed up the side of the housing in a "V" shaped space devoid of filters, which is bordered by filters. It first passes the filters next to the opening and can reach the filters diametrically opposite the inlet by passing around all of the filters in the middle of the housing.

Evidence that this channeling was occurring was obtained during leach tests of two filters which were removed for testing purposes (Table IV). The leaching solution in

TABLE IV

Concentration of Radium-226 in Leach Solutions

Description	226 Ra (pCi/l)
Cartridge No. 1 - Filter Batch	
3rd Circle of filters from outside, Mid-way from water input	
15 min. Leaches	
Leach 1 - pH 3.0 at start	16 ± 10
Leach 2 - pH 2.0 at start	122 ± 20
Leach 3 - pH 1.0 at start	625 ± 44
Leach 1M HNO_3	4150 ± 100
Cartridge No. 2 - Filter Batch 1	
1st (outside) Circle, 7/8 from input	
Leach - 24 hours @ 0.5M HNO_3	395 ± 9
Cartridge No. 3 - Filter Batch 2	
3rd Circle from outside, at input	
Not Leached	
Cartridge No. 4 - Filter Batch 2	
4th Circle, Mid-way from input	
Leach - 15 min. @ 0.5M HNO_3	2480 ± 80

each case was 0.5 M HNO_3, while the amounts of dissolved radium obtained from leaching differed by a factor of 6. The filter from which the higher amount of radium was removed came from near the middle of the housing, while the other filter came from diametrically opposite the input of the filter.

To re-route the flow of the incoming water, a baffle of corrugated iron sheet was cut to fit into the "V" shaped inlet slot and to act as a riser to allow water to flow up through the circular hole in the filter hold-down plate, around the edges of the plate and into the filter spring area, thence to the filters. The baffle was installed on January 18, 1979 but test results (See Table III) did not show any significant improvement. Whatever channeling or other preferential flow existed, there was no significant improvement over the radium removal rate before the baffle was installed.

Consideration was given to using a plugging medium to partially block some of the filters. This would tend to distribute water more evenly throughout the filter housing. However, this was not attempted due to perceived difficulties in operations.

At times it is desirable to backflush the filter housing, and some slight back flushing would occur with the well cycling on and off automatically during operations. This would dislodge any filter plugging medium. Suggested items to attempt the plugging were polyethylene sheet chips, sand, and diatomaceous earth.

During the project, a special apparatus for testing a single filter was developed. These tests were designed to determine the effect of varying the flow rate through the filters. Two separate tests were conducted. The second test was necessary due to the large variation in the radium content of the input water

which put the accuracy of the results of the first test in question.

With well 6 in operation, approximately 3 liters of water per minute are passed per filter. With well 7 added to the flow of well 6, this increases to around 3.5 liters per minute. With well 7 alone, the flow is about 0.5 liter per minute.

The results obtained are shown in Table V. At relatively low flow rates, the radium removal efficiency is somewhat greater than at higher flow rates; the same effect is also seen when Table III is examined. Since well 7, when used alone, produces water at a significantly slower rate than well 6, one would expect that the radium removal rates would be higher than when well 6 was used. This is observed. There is one apparent exception. In August of 1979, samples were collected of well 6, well 7 and well 6 plus well 7, both before and after the filter. Because insufficient time elapsed between well switch manipulations to allow the system to be purged of water from the previous arrangement, samples labeled well 7, and well 6 and 7, actually were of water which was at least partly of well 6, well 7 and/or wells 6 and 7. The volume of the housing was calculated to be about 250 gallons, and inasmuch as well 7 pumped only about 25 - 30 gallons per minute, about 10 minutes of operation would have been necessary for one filter housing volume to be processed (disregarding the volume of water in the lines). Since this was on the order of the length of time between samples in this particular series of samples, the obviously anomalous results were obtained.

There may well have been some preferential flow through the housing even with the baffle inserted in the housing, as the filters offered little resistance to the flow of water at the processing rate used (up to 120 gallons per minute).

Differential pressure across the housing was less than two pounds per square inch. It is thought that a higher differential pressure across the filters would lead to less uneven flow through the housing.

Based upon the data and experiments, it still cannot be discounted that significant preferential flow through the housing exists.

-3-

The first filter housing was not epoxy coated at the factory. It was thought that rust in the housing would not be significant, since some pipes in the city system had been in use nearly 50 years. Substantial amounts of rust were encountered, however. The chlorination step used for sterilizing the filters before filtered water could be added to the system produced water discolored with iron, and rusting was evident on the interior of the vessel and especially on the hardware (springs, "V"-bar guides, cups). The possibility that the iron from the vessel in the chlorine solution could somehow interfere with the ability of the manganese fibers to remove radium could not be discounted, so the interior of the vessel, the springs, cups, and "V" bars were wire brushed and painted with epoxy paint designed for painting the interior of water supply tanks. The filters from this first part of the test were removed and the second batch of filters were used for the second part of the test.

No effect of the iron could be demonstrated.

-4-

The fourth possibility, that of a filter inadvertently left out or a cap being knocked loose, causing a substantial volume of water to flow through the housing unfiltered was proven impossible by a careful inspection and count during the openings of the vessel.

The possibility that the first batch of filters, which contained significantly less manganese (about 6 - 7%) than the second (7 - 8%) was considered a likely cause of the low efficiency of the filter. The second batch did not perform as well as was expected from our earlier tests either.

It is quite likely though, that the effect of preferential flow in the single stage filter is a contributor to reducing the efficiency.

Our earlier bench-scale experiments used a two-stage filtration system. It was known that a higher filtration efficiency could be obtained at a lower flow rate per filter cartridge, and it was thought that the simplest way to apply the manganese fiber radium removal procedure would be to use a large single filter housing having a low flow rate per filter. In view of the low efficiencies actually found, it would appear that the best method would be to use at least two stages of filtration. This would allow the use of smaller units, and may circumvent the problem of waiting for a factory to custom make a larger unit, when the smaller ones may be more quickly obtained.

The use of smaller units would also result in a greater differential pressure across the unit and probably a more uniform flow through it.

VI. Recommendations for Use of Manganese Fibers to Routinely Treat Water for Removal of Radium

A. Planning

The process is at least 80% efficient in a single stage of filtration at first, and with two or more filter housings in series, should exceed 95%.

The single filter tests at various flow rates from 2 to 16 liters per minute, Table V, show a large filtering ability even at flow rates as high as 16 liters per minute (over four times the flow ratein the filter housing). Scaling down the size of the filter housing by a factor of 4 could be done and, assuming 80% radium removal each, for two housings in series the removal rate should be 96%.

PVC pipe fittings are less expensive and should be used. The installation is also simpler. They were avoided to prevent "false positives" in our tests for organic chemicals.

Treating used filters in potassium permanganate solutions after one use, instead of disposing of them directly and testing for radium removal ability, should also be tried.

Small filter housings are available and should be used. This would permit one person to perform filter handling operations, which due to the weight of the filter holddown plate, now requires two people to handle. The small filter housings also may be more readily available.

Table V

Single Filter Tests at about 3½ liters per Minute

Tests were performed to determine the

filter history on radium removal performance.

Cartridge Condition	Output pCi/l
Cartridge No. 3, Used, Not Leached	1.0 ± 1.5
Unused Filters	0 ± 0.2
Leached Cartridge	1.2 ± 1.6
Input water, well 6	29 ± 2.6

B. Operations

Step-by-step instructions for treatment, filter loading and disposal are included as Tables VI, VII, and VIII.

C. Costs

The costs of operation could not be determined as well as desired due in part to a reduction of state travel funds and the absence of the second filter housing. More data should be available for the final version of this report. The cost analyses shown below (Table IX) are therefore maximums. The cost total for expendable supplies and labor is $1,137.75 for one filter housing change (sufficient for at least 2.5 million gallons), or approximately 45 cents per thousand gallons (maximum). Laboratory analyses, overhead, amortization and installation of equipment costs are not included, nor is modification of the system to provide series filtration. More data on filter exhaustion would reduce costs for these items.

This analysis shows that this process can favorably compete with other water treatment processes. Any attempt to remove the radium and re-use the filters should serve to greatly lower the cost of new filters, (the major expense), but should only be undertaken by persons with a familiarity of health physics. Means for neutralizing and solidifying wastes generated should also be included.

Reprocessing the filter should also include a bath in a potassium salt, KCl for example, as it is suspected that the process operates as an ion exchange between K^{+} and Ra^{++}.

An efficient crew accustomed to installing equipment in water treatment plants should be able to conduct operations according to the schedule in Table X.

TABLE VI

Step-by-Step Instructions

- Filter Treatment -

For 120 Filters

Minimum crew: 1 (2 desirable)

Materials needed:

Work area (with water supply, drain, electricity)

2 - 55 gallon drums with lids

2 - 53 gallon 1/8" rigid polyethylene drum liners

*60 lbs. reagent grade potassium permanganate ($KMnO_4$) crystals

*120 - 30 inch spun acrylic filters

1½ mil polyethylene sheet - two 3' diameter circles

2 polyethylene buckets - 2 to 5 gallon size with pouring spout

Drum syphon pump

**Stirring stick

*70 gallons Hot (110 - 120°F) water

**2 - Drum heating strips, rings and 20 gauge wire (15')

2 - Drum heater variable power supplies

Polyethylene scoop (for $KMnO_4$)

50 foot garden hose connected to water supply

**Rubber gloves - dishwashing type

Rubber boots and rain suit (plastic, water repellant)

*24 Plastic bags - leaf type, sturdy (1.5 mil) minimum 36" depth

Paper towels

Goggles (for splash protection)

---

*Consumed, not reuseable

**Subject to rapid wear/or chemical attack

Table VI (Cont'd)

Remember - these filters are to be installed in your drinking water supply.

Keep them clean.

1. Read warning on chemical containers. Potassium permanganate, ($KMnO_4$) improperly handled, can cause fire and/or explosions.

2. Place drums in work area.

3. Put liners inside drums.

4. Put (clean) gloves on.

5. Place 60 filters in drum, arranging them vertically (it may be helpful to tip drum to a 45° angle).

6. Put boots and rain suit on.

7. Connect hose and turn on water. Leave water on!

8. Fill bucket approximately ½ full with cold water. Add warm water to fill bucket, adjust temperature of water bucket to 86 - 90°F (30 - 32°C).

9. Pour warm water from first bucket into second bucket (called chemical bucket) to fill half full.

10. Add up to 15 lbs. of $KMnO_4$ to water (carefully!) in chemical bucket (use scoop). Keep crystal level about 1/5 of bucket depth or less.

11. Stir gently for about 3 - 5 minutes. (Rinse off any spilled chemical solution immediately).

12. Pour chemical solution (but not any undissolved crystals) from chemical bucket carefully onto filters. Rinse off any spills.

13. Repeat steps 9 - 12 until chemical level comes up to within one inch of the top of the filters, keeping the water bucket temperature no warmer than 90°F.

14. Measure the temperature of the potassium permanganate solution in the drum. If it is below 86°F (30°C), add hot water to drum to bring the liquid level up to within 1½ inches (4 cm) of the lip of the liner.

15. Put syphon pump in drum.

16. Holding pump hose so that any pumped solution runs back into the drum, prime the pump. Lift the chemical bucket up and hold it over the chemical solution in drum (to avoid spilling on the floor), put the hose in the bucket and lower the bucket so that the syphon brings solution from bottom of drum into bucket. As the bucket fills to 2/3 full, stop the flow by opening the syphon relief valve.

17. Stir solution and crystal mixture in bucket for several minutes.

18. Pour solution into drum (same as step 12).

19. Repeat steps 16 - 18 until all crystals have dissolved.

20. Add hot water to bring liquid level in liner up so that filters are covered by at least 2 cm (3/4 inch) of solution. Filters may float, and will need to be pushed down.

21. By repeatedly filling and emptying bucket using syphon pump, cycle 50 gallons of solution from the bottom of the drum to the top to mix solution well.

22. Fill the small annular space between the drum and drum liner with water to within 3 inches of the top. This "jacket" of water conducts the heat from the drum heater to the filters.

23. Rinse off the drum and other equipment and dry drum with paper towels.

24. Wrap the drum heater around the base of the drum. Use the small spring and wire to keep it in good contact with the drum by wrapping the wire once around the drum. Connect the spring to one end of the heater, the wire to the other end of the spring and also to the other end of the heater strip. Spring should be stretched so that drum heater remains in good contact with the drum.

25. Place round polyethylene sheet cover on top of filters and cover with drum lid.

26. Connect heater power supply, and test for operation. Allow 8 - 16 hours for stable temperature to be reached. Adjust the power supply to keep within the range 86°F (30°C) to 104°F (40°C).

27. Once each day or two, repeat Step 21. Remove the heater before working with solutions. Replace heater afterwards.

28. After 2 or 3 days, the transparency of the solution will begin to change from a dark, deep purple to a more transparent and lighter shade. Some brown fine precipitate (manganese oxide) will also form.

    As soon as any noticeable lightening in color has occurred, add 5 pounds of $KMnO_4$ to the solution in the drum by disconnecting the drum heater and removing the heater strip (to keep accidental spills off of it) and following these steps:

29. Connect hose and turn on water and <u>leave on</u>.

30. Put syphon pump in drum.

31. Holding pump hose so that any pumped solution runs back into the drum, prime the pump. Lift the chemical bucket up and hold it over the chemical solution in drum (to avoid spilling on the floor) put the hose in the bucket and lower the bucket so that the syphon brings solution from bottom of drum into bucket. As the bucket fills to 2/3 full, stop the flow by opening the syphon relief valve.

32. Add 5 lbs. of crystal potassium permanganate to the bucket and stir.

33. Stir solution and crystal mixture in bucket for several minutes.

Table VI (Cont'd)

34. Pour solution into drum (same as step 12).

35. Repeat steps 31, 33 and 34 until all 5 lbs. have been dissolved.

36. Reconnect heater, following steps 23, 24, 25 and 26.

37. Twice more over the next few days, as the strength of the solution decreases, add more $KMnO_4$ in 5 lb. lots following steps 28 through 36.

After all these steps have been completed and the solutions kept warm, the purple color will completely disappear, indicating that all 30 pounds of $KMno_4$ in each drum has been converted to oxides of manganese and potassium. Once the color of the liquid has changed to clear, the heaters may be disconnected and removed. The filter fibers should contain about 7.5% Manganese by weight. When the brown precipitate is washed off of the filters they will have a very dark chocolate to black color. Check one or two of the filters by removing and washing them. The filters are ready for loading into the filter housing now.

Keep the filters in the solutions without rinsing, until they are ready for use.

TABLE VII

Step-by-Step Instructions

Loading Filter Housing

Minimum Crew: 2

Materials:

1 quart bottle 5% liquid chlorine bleach

Boots, rain suits, gloves

24 plastic garbage bags (needed only if transportation of filters is necessary)

115 treated filters

Wrenches and jack for filter housing opening

Paper towels

Hose connected to water system

Filter housing internal parts

Procedure

1. Open bypass valve and close inlet and outlet valves on housing
2. Open drain valve. Open filter housing and insert rods for filters in cups in bottom of housing.
3. Put on gloves, boots and rain gear.
4. Remove filters from lined drums (place 5 in each bag if bagging).
5. Slide filters over rods onto cups until all 115 filters are in place. Fill from the center outward.
6. Count the number of filters installed. (Every one of the 115 filter positions must be filled).
7. Place cups with springs on the filters, beginning in the center and moving outward.
8. Lift the cover plate over the springs, align holes and slide over vertical bolts. (This must be done carefully to avoid dislodging springs). If any springs fall into the housing, it will be necessary to retrieve and replace them. A straightened wire from a coat hanger and a flashlight will be of great assistance.

Table VII (Cont'd)

9. Tighten bolts to hold filters in place.

10. Supply water pressure to the filter inlet valve.

11. Open filter inlet valve and allow water to run into filter housing and overflow. Water will also exit through the opened housing drain valve.

12. Close filter inlet, allow water level in housing to drop to about 1/3 of the length of the filters and close the filter drain.

13. Put rubber gasket in place and prepare to lower filter housing head.

14. Pour the entire bottle of bleach into filter housing through the hole in the hold-down plate over the inlet "V".

15. Lower and bolt down the head of the housing.

16. Open filter housing inlet valve slightly. Air will be expelled through the air relief valve in the top of the housing as the housing is filled. When the sound of the air flowing out of the valve ceases, the housing is full.

17. Open the filter housing drain valve fully for a few seconds, and close it when the smell of the bleach is detectable.

18. Close the filter inlet valve tightly. Pressure to the housing inlet valve now may be removed if necessary.

19. IMPORTANT: The filter housing must remain undisturbed, full of the chlorine solution for at least 24 hours for proper disinfection.

20. After at least 24 hours, open the filter housing drain valve and let housing empty. Smell the water to verify a strong chlorine bleach smell is still present.

21. Apply water pressure to the filter input valve.

22. Open the inlet valve on the filter housing, letting water flow from the main, through the filters and then waste the water out of the filter drain valve to purge the filter housing of chlorine and precipitated manganese.

23. Purge the housing of this by occasionally shutting off and opening the drain valve.

24. After no further brown precipitate is seen, leave the filter drain valve open and close the filter by-pass valve. The water in the main will now flow through the filter and be dumped through the filter drain valve.

25. The filter output valve may now be opened to feed treated water into the system. Close filter drain valve.

Table VII (Cont'd)

26. Close the filter drain valve and regulate filter pressure. (30 psig for well 6 and 7 - 25 psig for well 6 or 7 alone)

TABLE VIII

Step-by-Step Instructions

Filter Removal and Disposal

Minimum Crew: 2

Materials:

Hose connected to water system

24 plastic garbage (leaf) bags minimum height 36"

2 drums for radioactive waste with absorbent material.

Disposable gloves, booties, overalls (water repellant)

Portable alpha survey meter

Masking tape

Paper towels

Box (for springs) - 1 cubic foot size

Wrenches and jack for opening housing

Dishwashing detergent

Procedure

1. Open bypass valve if water needed in system
2. Drain filter housing by closing intake and outlet valves, and opening low point 1" valve on bottom of housing. Allow at least 24 hours for filters to drain.
3. Open drums, put plastic liner in, drape excess over drum lip and side. Put 1" of absorbent material on bottom of drum. Tip drum to about 45° angle to facilitate loading.
4. Put on disposable gloves, booties and rain suit. Tape sleeves.
5. Open housing, remove spring cover plate, remove spring/cap assembly on filters.
6. With a helper holding the plastic bag, remove 5 filters, one at a time, and put them in the plastic bag. Tie bag. Helper puts the bag in the drum.
7. Repeat step 6 until 60 filters have been put in the first drum and 55 in the other.
8. Put disposable gloves, shoes, towels and coveralls from the last job in the small empty space in the side of the second drum, add absorbent material on

top of the filters, tie drum lining bag and seal drums. Wipe off drums with towels. Remove protective clothing. Put coveralls, gloves, booties, and towels in plastic bag to be stored for disposal at the next filter unloading. Survey personnel for contamination (hands, face, clothes, feet) using alpha survey meter.

9. Survey drum for:

gamma ray radiation levels

Alpha contamination

Wipe drum with filter paper for contamination test.

Count wipe (must be less than 22,000 dpm) to meet U. S. Department of Transportation Regulations. Should not exceed 500 dpm.

10. Using damp paper towels, wipe off any area of skin which shows any evidence of foreign material from the filters. Using liquid dishwasing detergent, wash areas. Discard towels in plastic bag to be saved for disposal at next replacement.

11. Perform final decontamination survey on personnel. Remove any residual contamination by washing. Skin must be dry to detect alpha contamination.

12. Call disposal company to pick up drum.

Table IX

Costs of Operation

Consumable Supplies per Filter Loading

Filters	115 @ $7.05	810.75
Potassium permanganate	60 lbs. @ $2.45	157.00
Miscellaneous		10.00
Sub Total		$977.75
Labor	16 hrs. @ $10.00	160.00
Total		$1,137.75

Table X

Time Analysis

(Person Hours)

			Total Hours
Filter Housing Installation			
Foundation	2 people	6 hours	12 hours
Plumbing	3 people	10 hours	30 hours
Filter Preparation			
Initial Loading Chemical Mixing	2 people	4 hours	8 hours
Checking, Stirring, Mixing	1 person	8 hours	8 hours
Filter Installation			
Transportation	1 person	½ hour	½ hour
Vessel Opening	2 people	¼ hour	½ hour
Installation	2 people	1 hour	2 hours
Vessel Closing and Chlorination	2 people	½ hour	1 hour
Flushing	1 person	½ hour	½ hour

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